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Freestanding and flexible graphene paper as support for biocatalysts in enzymatic biofuel cells

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Biocatalysts for enzymatic biofuel cells (EBFCs) require more practicable and spendable electrode materials for accommodation of the enzymes than the traditional solid graphite or expensive noble metal electrodes. We report here the use of novel graphene paper (GP) as a freestanding and flexible cathode and anode electrode material for immobilization of enzymes in a glucose/O_2 EBFC. GP electrodes were prepared via controlled assembly of graphene oxide (GO) nanosheets into a freestanding structure, followed by the specific reduction of GO to reduced GO (rGO). Bilirubin oxidase (BOx) physically adsorbed on the GP electrode, can directly transfer electrons between BOx and the cathode, facilitating the dioxygen reduction reaction (ORR). Likewise, pyrroloquinoline quinone dependent glucose dehydrogenase (PQQ-GDH) was physically immobilized onto a GP anode. However, direct electron transfer between PQQ-GDH and the anode cannot occur because the active site of the enzyme is deeply buried in the protein structure. Meldola blue (MB) was therefore introduced as a mediator shuttling electrons between PQQ-GDH and the anode, and facilitating PQQ-GDH catalysed glucose oxidation. As a result, an EBFC with a robust open circuit voltage up to 0.625 V and max power density of 3.97 $\mu W/cm^2$ could be obtained. These values are competitive with established EBFCs. The new GP supported EBFC holds potential for operating under *in vivo* conditions, e.g. integrated in biological compartments, and with minimal risk of blood clotting and plaque formation, and driving a low-power device such as a pacemaker.